REMARKS

This application amends claims 12, 15 and 22, and adds new claims 34-36. Support for the claim amendments and new claims can be found in the specification as originally filed. No new matter has been added. Claims 12-36 are pending in the application.

Claims 12-33 stand rejected under 35 U.S.C. §103(a) for obviousness over U.S. Patent No. 6,290,913 to Aoyama.

Amended independent claim 12 is directed to a system for removing carbon monoxide from a hydrogen-containing treatment-object gas containing hydrogen as its major component as well as carbon dioxide. The system includes two stages of CO removers for removing carbon monoxide. The first stage CO remover removes a portion of the carbon monoxide by methanation through a catalyst reaction by a first metal catalyst. The catalyst comprises one or more selected from the group consisting of Ru, Pt, Rh, and Pd. The first stage CO remover is capable of methanating carbon monoxide through a catalyst reaction for methanating the carbon monoxide while restricting methanation of the carbon dioxide. The second stage CO remover removes a remaining portion of the carbon monoxide mainly by oxidation through a further catalyst reaction involving addition of an oxidizing agent.

Amended independent claim 15 is directed to a method of removing carbon monoxide from a hydrogen-containing treatment-object gas containing hydrogen as its major component as well as carbon dioxide. The method includes a first step of causing the treatment gas to contact a first metal catalyst comprising one or more selected from the group consisting of Ru, Pt, Rh, and Pd and capable of methanating carbon monoxide at a temperature where methanation of carbon monoxide takes place by the first metal catalyst. A portion of the carbon monoxide is thus removed through carbon monoxide methanation and methanation of the carbon dioxide is restricted. The second step of causing the treatment-object gas from the first step includes an oxidizing agent to contact a second metal catalyst capable of oxidizing carbon monoxide so that a remaining portion of carbon monoxide is removed mainly through carbon monoxide oxidation.

Aoyama discloses an apparatus and method for reducing the concentration of carbon monoxide in a carbon monoxide containing gas. Fig. 22 of Aoyama discloses a methanization unit (94) arranged after a CO-selective oxidizing unit (34). The reformed gas discharged from the CO-selective oxidizing unit (34) is subjected to the methanization

reaction of carbon monoxide in the methanization unit (94) before being supplied to the fuel cells. In another embodiment shown in Fig. 26, Aoyama discloses a methanization reaction occurring simultaneously with the selective oxidation reaction of carbon monoxide.

Aoyama does not teach or suggest a system or method of removing carbon monoxide from a hydrogen-containing treatment-object gas as recited in amended independent claims 12 and 15. In particular, Aoyama does not teach or suggest a system or method having a methanization stage/step prior to a second stage/step of oxidation for removing carbon monoxide from a hydrogen-containing gas. The system and method of the present claimed invention achieves a low final carbon monoxide concentration, as well as provides minimization of hydrogen consumption. For example, the present invention is directed to a carbon monoxide removing technique capable of effectively reducing/removing carbon monoxide present at 1,000 ppm to several percent in a hydrogen-rich treatment gas to a concentration of several tens of ppm (such as 10 ppm or lower). Additionally, the present claimed invention achieves minimization of hydrogen consumption and the possibility of CO concentration to a desired near-lower limit level under the condition of co-existence of carbon dioxide. The below illustration establishes the minimization of hydrogen consumption achieved in the present claimed invention.

An analysis for a treatment-object gas containing 1% CO will be compared when removing carbon monoxide by (a) an oxidation reaction alone, (b) a methanation reaction alone, (c) an oxidation reaction, followed by a methanation reaction as set forth in Aoyama, and (d) a methanation reaction followed by an oxidation reaction as in the present claimed invention.

(a) <u>oxidation reaction alone</u>

In this case, an oxidizing agent, for example air, is inputted to the treatment-object gas for oxidation of CO. In this, the oxidation reaction of carbon monoxide and the oxidation reaction of hydrogen proceed simultaneously.

Further, as pointed out by the Applicants on page 3, lines 1-5 of the application, and also in column 17, lines 4-43 of Aoyama, the molar ratio of oxygen to carbon monoxide is 3, of which 0.5% is consumed for the oxidation of the 1% CO whereas the remaining 2.5% is consumed for the oxidation of hydrogen. Therefore, <u>5%</u> of the hydrogen is consumed during the process of an oxidation reaction alone.

(b) methanation of carbon monoxide alone

In this instance, if the methanation reaction takes place selectively, the production of methane and water is involved. Therefore, 3% of hydrogen is required and consumed for the 1% CO. Furthermore, this process does not allow a CO concentration to be reduced to 10 ppm or lower, which is the preferred range of the present invention.

(c) oxidation reaction followed by methanation reaction

Aoyama is directed to this process of oxidation followed by a methanation reaction. In this instance, if the oxidation process removes 70% of CO and the methanation step removes the remaining 30% of the CO, the total amount of hydrogen consumed is: (5 x 0.7) + (3 x 0.3) = 4.4. Therefore 4.4% hydrogen is consumed in total during this process.

(d) methanation reaction followed by oxidation reaction

The present claimed invention as recited in independent claims 12 and 15 utilizes a methanization step prior to oxidation. If the first-stage CO remover (methanation) removes 70% of CO and the second-stage remover (oxidation) removes the remaining 30% of CO, the total amount of hydrogen consumed is: (3 x 0.7) + (5 x 0.3) = 3.6. Therefore, there is a consumption of 3.6% hydrogen in total.

Thus, as herein illustrated, the reaction order utilized in the present claimed invention, namely a methanation reaction followed by an oxidation reaction, is more advantageous for a reduction of the total amount of hydrogen consumed and required for CO removal. Clearly, the process of Aoyama discloses a higher hydrogen consumption than that of a methanation step prior to oxidation in the present claimed invention. Hence, contrary to the Examiner's statement on page 4 of the Office Action, one skilled in the art concerned with hydrogen loss in a hydrogen-rich gas would not utilize the process as disclosed in Aoyama.

Additionally, the present claimed invention achieves a greater energy balance than that of the process disclosed in Aoyama. A methanation reaction and an oxidation reaction respectively involve thermal energy generation, in other words, consumption of energy available from the fuel whose amounts are calculated as shown below.

oxidation reaction

$CO + \frac{1}{2} O_2 \rightarrow CO_2$ $\Delta H^0 = - 283.0$ kJ/mol

$H_2 + \frac{1}{2} O_2 \rightarrow H_2O$ $\Delta H^0 = - 241.0$ kJ/mol

methanation reaction

$CO + 3H_2 \rightarrow CH_4 + H_2O$ $\Delta H^0 = -205.7$ kJ/mol

$CO_2 + 4H_2 \rightarrow CH_4 + 2H_2O$ $\Delta H^0 = -164.5$ kJ/mol

With these reactions in consideration, the above-described two cases (c), (d) involving the combination of these reactions will be analyzed on the same assumption as used above with hydrogen consumption, the first-stage being able to remove 70% CO and the second stage being able to remove 30% CO.

(c) oxidation reaction followed by methanation reaction

CO: 1 mol $\rightarrow$ CO: 0.3 mol $\rightarrow$ CO: 0 mol

(-283.0 x 0.7) + (-205.7 x 0.3) = -259.81 kJ

(d) methanation reaction followed by oxidation reaction (present invention)

CO: 1 mol $\rightarrow$ CO: 0.3 mol $\rightarrow$ CO: 0 mol

(-205.7 x 0.7) + (-283.0 x 0.3) = -228.89 kJ

If processes (c) and (d) are compared, without regard to the side reactions in the oxidation and the methanation steps, it is established that the discharged heat loss is smaller in the process of the claimed invention, (d), -228.89 kJ than the process recited in Aoyama, (c), -259.81 kJ as illustrated above. Hence, the thermal energy preserved in anode off gas is greater, thus achieving a higher energy consumption efficiency in a system in which the off gas is recycled. Furthermore, even when the respective side reactions in the oxidation reaction and the methanation reaction occur in the above processes, the reaction heat amount -164.5 kJ/mol in the CO_2 methanation reaction as the side reaction of the CO methanation step is smaller than that (-241.0 kJ/mol) in the H_2 oxidation as the side reaction of the CO oxidation step. Therefore, this further evidences that the discharged heat loss due to the side reaction is also smaller in (d) than (c).

Moreover, the above is further appreciated when considered with respect to a fuel cell system as defined in new claims 35 and 36 having a CO removing system of the present claimed invention. Assuming that this fuel cell system receives a fixed amount of fuel (i.e. total energy input being fixed), it is more advantageous to preserve the energy in the methane production and to reuse it. For instance, by reusing the methane produced in the first-stage CO remover within the system, efficient utilization of energy can be achieved. This is not possible for carbon dioxide.

Additionally, although it is easier to achieve CO concentration reduction to 10 ppm with oxidation of CO than with methanation of CO, if CO is removed only by the oxidation, this will result in a large consumption of an oxidizing agent which consequently results in a large amount of hydrogen fuel consumption. As discussed above, to minimize hydrogen consumption, the methanation of CO occurs first so that some of the hydrogen may remain in the form usable as a fuel, i.e. for production of methane, while the CO concentration is reduced as much as possible. Then the oxidation removal occurs on the remaining CO concentration with the addition of a small amount of another oxidizing agent on the remaining CO which has been reduced already to a relatively low concentration in the preceding first stage. Accordingly, the invention achieves the above object with the CO concentration reduced to an order of several ppm or lower. In contrast, with the construction of oxidation first and then methanation as described in Aoyama, it is difficult to achieve such a low final CO concentration without a large amount of hydrogen loss due to the production of water, wasteful energy consumption, methanation of carbon dioxide, etc.

Moreover, the conventional construction described in Aoyama of "oxidation followed by methanation" can be useful or appropriate in a system where the energy utilization ratio can be relatively low, as in the case of a system for automobiles. On the other hand, the construction of the present claimed invention "methanation followed by oxidation" allows for efficient utilization of energy available from the fuel and, therefore, can be readily used in systems such as a stationary (ground-installed) system. For all the foregoing reasons above, Aoyama clearly does not teach or suggest reducing CO concentration in a hydrogen-rich gas by methanation prior to oxidation. Applicants respectfully request reconsideration of amended independent claims 12 and 15.

Claims 13, 14, and 21; and 16-20 and 22-33 depend directly or indirectly from and add further limitations to amended independent claims 12 and 15, respectively, and are believed to be patentable for all the reasons discussed above in connection with amended independent claims 12 and 15. Reconsideration of the rejection of claims 13, 14, and 21, and 16-20 and 22-33 is respectfully requested.

New Claim 34 depends from and adds further limitations to amended independent claim 15 and is believed to be patentable for the same reasons discussed hereinabove in connection with amended independent claim 15.

Claims 35 and 36 are newly added. Independent claim 35 is directed to a fuel cell system including a carbon monoxide system as recited in independent claim 12. The methane produced by the first–stage CO remover is utilized as a reforming fuel. Independent claim 36 is directed to a method of operating a fuel cell system. The method includes implementing a carbon monoxide removing method as disclosed in amended independent claim 15 and using the methane produced at the first step as a reforming fuel.

New independent claims 35 and 36 are believed to be patentable for the same reasons discussed hereinabove in connection with amended independent claims 12 and 15. Aoyama fails to teach or suggest a fuel cell or method of operating a fuel cell as recited in independent claims 35 and 36 with a first-stage/step removal of CO by methanation and a second step/stage removal of CO by oxidation and using the methane produced at the first step as a reforming fuel.

Based on the foregoing amendments and remarks, reconsideration of the rejections and allowance of pending claims 12-36 are respectfully requested.

Respectfully submitted,

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